

Optical Rotation Quasi-Phase-Matching for Circularly Polarized High Harmonic Generation

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The first scheme for quasi-phase-matching high harmonic generation of circularly polarized radiation is proposed: optical rotation quasi-phase-matching (ORQPM). In ORQPM propagation of the driving radiation in a system exhibiting circular birefringence causes its plane of polarization to rotate; by appropriately matching the period of rotation to the coherence length it is possible to avoid destructive interference of the generated radiation. It is shown that ORQPM is approximately 5 times more efficient than conventional QPM, and half as efficient as true phase-matching.

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When an intense laser pulse on the order of 10^{14} W/cm² is focussed into a low density gas high order harmonics of the fundamental driving field can be produced. This high harmonic generation results in coherent radiation extending to soft x-ray wavelengths. As such it is an attractive source of EUV radiation and has applications including time-resolved science [1], ultrafast holography [2], and coherent diffractive imaging [3].

However, without additional techniques, HHG is very inefficient; typical photon conversion efficiencies are 10^{-6} for generated photons with energies of order 100 eV, decreasing to 10^{-15} for generation of 1 keV radiation. This inefficiency is largely due to the fact that the driving and harmonic fields have different phase velocities; as such they develop a phase mismatch, causing the intensity of the generated harmonics to oscillate with propagation distance with a period $2L_c = 2\pi/\Delta k$. The wave vector mismatch Δk is in general non-zero owing to dispersion in the target material, geometric dispersion, and waveguide dispersion; it is given by $\Delta k = k(q\omega) - qk(\omega)$, where $k(\omega)$ is the wave vector of radiation of angular frequency ω and q is the harmonic order.

The efficiency of HHG can be greatly increased by true phase-matching, i.e. balancing dispersion in the system so that $\Delta k = 0$. In the case of harmonics generated in a hollow core waveguide true phase-matching may be achieved by tuning the pressure in the waveguide, enabling the quadratic growth of the harmonic intensity with propagation distance. [4]. However, true phase-matching may only be achieved up to a critical ionization level, above which it is no longer possible to achieve $\Delta k = 0$, placing a limit on the maximum harmonic order which can be phase-matched. An alternative approach to improve the efficiency of HHG is to employ the technique of quasi-phase-matching, although QPM is not as

efficient as true phase-matching. In QPM, HHG is suppressed in regions where the locally generated harmonic is out of phase with the harmonic beam. By suppressing HHG in multiple out-of-phase regions the harmonic intensity grows monotonically with z . Techniques for QPM include the use of counter-propagating pulses [5], multi-mode beating [6], modulated waveguides [7], and modulated gas density [8], and static electric fields [9].

Recently we proposed a new QPM technique: polarization beating QPM (PBQPM) [10,11]. In this approach, a linear birefringent system modulates the polarization of the driving pulse, causing it to beat between linear and elliptical. Because harmonic generation is suppressed for elliptically polarized light, QPM can be achieved if the period of the polarization beating is suitably matched to the coherence length. In this paper, we propose a novel, more efficient, QPM scheme that enables the generation of circularly polarized high harmonics: Optical Rotation Quasi Phase Matching (ORQPM) [12]. ORQPM utilizes a waveguide with circular birefringence (as opposed to linear birefringence), which causes the plane of polarization of linearly polarized light to rotate with propagation distance at a constant rate, with period $2L_r$. By matching $L_r = L_c$, the generated harmonics will grow monotonically. As we show in detail below, ORQPM allows harmonics beyond the true phase-match limit to be generated with comparable efficiencies to that obtained with true phase-matching. Moreover, ORQPM is the first QPM scheme to generate circularly polarized high harmonics; bright sources of circularly polarized soft x-radiation would find widespread application in studies of ultrafast spin dynamics [13] and nano-lithography [14]. We also note that a polarization gating technique in metallic macrostructures has been proposed to generate circularly polarized high harmonics [15] and that second

harmonic generation quasi-phase matching has been previously achieved in an optically active solid media [16]. In this paper we describe ORQPM in detail, demonstrate its operation by means of a simple model, and discuss three techniques by which it might be realized.

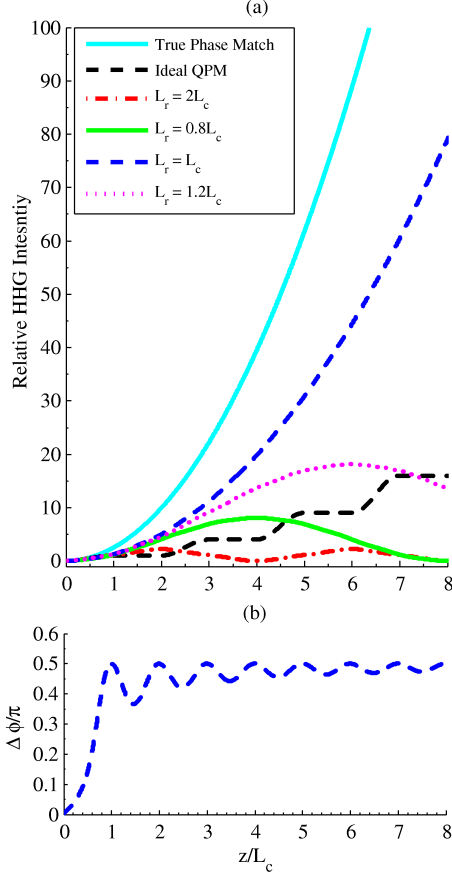


Fig. 1. (a) Relative HHG intensity for true phase matching (solid cyan line), ideal QPM (dashed black line), and ORQPM for $L_r = 2L_c$ (dot-dashed red line), $0.8L_c$ (solid green line), L_c (dashed blue line), $1.2L_c$ (dotted pink line) normalized to the the output after a single coherence length with no phase matching. (b) Phase difference $\Delta\phi = \arg(\hat{\xi}_x) - \arg(\hat{\xi}_y)$ between the x and y polarization components as a function of z for $L_r = L_c$.

The driving electric field can be written as a linearly polarized wave with a plane of polarization that rotates with propagation along the z axes with period $L_r = \pi/\nu$, where ν is the optical rotation power:

$$\vec{E}_{\text{driver}}(z, t) = E_0 e^{i(\beta z - \omega t)} \begin{pmatrix} \cos(\nu z) \\ \sin(\nu z) \end{pmatrix}. \quad (1)$$

where $\beta = k(\omega)$ is the propagation constant and E_0 is the field amplitude. For the purpose of demonstrating ORQPM, we will assume that the damping of the driving radiation is negligible.

In the slowly-varying envelope approximation, the electric field of the q th harmonic can be written as:

$$\vec{E}_q(z, t) = \vec{\xi}(z, t) e^{i[k(q\omega)z - q\omega t]} \quad (2)$$

where $\vec{\xi}$ is the electric field envelope. The Rayleigh range of the harmonic beam will be approximately q times that of the driving beam; as such we will ignore the effect of the waveguide on the wave-vector $k(q\omega)$ of the harmonic beam. We note that in cases where $k(q\omega)$ is affected by the waveguide, ORQPM will still occur if there is a difference between the circular birefringence experienced by the driving and generated beams.

Under a continuous-wave approximation, the amplitude of the x - and y - components of the harmonic envelope will grow according to,

$$\begin{cases} \frac{\partial \xi_x}{\partial z} = A \Lambda_x(z) e^{-i\Delta k z} \\ \frac{\partial \xi_y}{\partial z} = A \Lambda_y(z) e^{-i\Delta k z} \end{cases} \quad (3)$$

where A is a normalization constant, and Λ_x and Λ_y are the relative source terms. In this case the driving radiation has the same intensity at all points along the system, and is always linearly polarized. The locally generated harmonics will have a polarization parallel to that of the driving beam, and hence $\Lambda_x = \cos(\nu z)$ and $\Lambda_y = \sin(\nu z)$.

Assuming that A and Δk are constant, Eqn. (3) yields,

$$\begin{cases} \xi_x(z) = iA \frac{\Delta k - e^{i\Delta k z} [\Delta k \cos(\nu z) + i\nu \sin(\nu z)]}{\Delta k^2 - \nu^2} \\ \xi_y(z) = A \frac{-\nu + e^{i\Delta k z} [\nu \cos(\nu z) + i\Delta k \sin(\nu z)]}{\Delta k^2 - \nu^2} \end{cases} \quad (4)$$

ORQPM corresponds to setting $\nu \rightarrow \Delta k$, whereupon,

$$\begin{cases} \hat{\xi}_x(z) = \lim_{\nu \rightarrow \Delta k} [\xi_x(z)] = A \frac{2\Delta k z - i + i e^{-2i\Delta k z}}{4\Delta k} \\ \hat{\xi}_y(z) = \lim_{\nu \rightarrow \Delta k} [\xi_y(z)] = A \frac{-2i\Delta k z + 1 - e^{-2i\Delta k z}}{4\Delta k} \end{cases} \quad (5)$$

From this the intensity, $\hat{I}(z) = \hat{\xi}_x \hat{\xi}_x^* + \hat{\xi}_y \hat{\xi}_y^*$ is found to be:

$$\hat{I}(z) = A^2 \left[\frac{1}{2} z^2 + \frac{1 - \cos(2\Delta k z)}{4(\Delta k)^2} \right]. \quad (6)$$

We see that the growth of the harmonic intensity comprises a quadratic term, which dominates at large z , plus a weak co-sinusoidal modulation. It is clear that in the limit of large z , ORQPM is half as efficient as would be true phase-matching — i.e. setting $\Delta k = 0$ — under otherwise identical conditions.

These analytical results are confirmed by the results of numerical integration of Eqn. (3), as shown in Fig. 1. When properly matched, ORQPM causes the intensity of the harmonic to grow almost monotonically, and it may be seen that at large z that the intensity is half that which would be obtained with true phase-matching. Notice that with ORQPM the harmonic intensity grows $\pi^2/2 \approx 5$ times faster than ideal QPM, defined to be the square-wave modulation of the local harmonic generation with a period $2L_c$ and complete suppression of harmonic generation in the out of phase zones.

In Fig. 1(b) the phase difference between the two polarization states of the harmonic is illustrated; it is seen that after a few coherence lengths this phase difference is close to $\pi/2$, corresponding to circular polarization of the harmonic. This can also be seen in Eqn (5), where the

dominating terms for the x - and y - components of the envelope function, $\frac{A}{2}z$ and $-\frac{iA}{2}z$ respectively, have the same amplitude but are $\pi/2$ out of phase, corresponding to circular polarization. For fixed t , the rotation with z of the plane of polarization of the harmonic radiation is in the same sense as that of the driving beam, i.e. right(left)-hand polarized harmonics are generated by right(left)-rotating driving radiation.

In order to achieve ORQPM in practise it would be necessary to guide a laser pulse over an extended region using a waveguide with circular birefringence that has a rotation length shorter than 1mm. Such a waveguide could be constructed in a number of ways. One possibility for achieving ORQPM is a waveguide constructed from a material with a high Verdet constant V , which would allow ORQPM to be achieved by tuning the applied magnetic flux density B until $\nu = VB = \Delta k$. We note that Verdet constants lie in the range $100 \text{ deg mm}^{-1} \text{ T}^{-1}$ to $10^5 \text{ deg mm}^{-1} \text{ T}^{-1}$ [17]. Solid-core waveguides exhibiting Faraday rotation have been developed, although the hollow-core systems which would be required for ORQPM have yet to be developed [18, 19].

Alternatively, the waveguide walls could be constructed from optically active materials, which have rotary powers in the range 10^2 to 10^4 deg mm^{-1} [20, 21]. Further, we note that polarization rotating photonic crystal fibres have been developed with $L_r < 1\text{mm}$ [22] and that HHG has been achieved using PCFs [23].

Finally, we also note that ORQPM could be achieved in non-birefringent waveguides by exciting two circularly polarized waveguide modes with different mode velocities. The resultant superposition of these two modes will be linearly polarized with rotating polarization.

As for any QPM scheme limits will be set by absorption of the harmonics and variation of the coherence length [24]. As for other QPM schemes based on polarization-control of the driving laser, the distance over which ORQPM can be achieved may be limited by relative slippage of the constituent polarizations. However, we note that in principle this limit can be avoided by reversing the rotation of the driving field when the two modes have slipped apart, although the polarization of the resultant harmonic field may no longer be perfectly circularly polarized. Finally, we point out that ORQPM may also be limited by chromatic dispersion from either waveguide or Faraday effects.

In this paper, we have proposed a novel QPM scheme for high harmonic generation that relies on rotation of the driving radiation using a circularly birefringent waveguide. By matching the rotation length to the coherence length, circularly polarized harmonics may be produced with an efficiency up to 50% of that obtained with true phase-matching and 5 times more efficient than conventional QPM. These findings were confirmed by numerical simulations. Three systems in which ORQPM might be achieved have been identified; exploration of these will form the basis of future work.

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